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Deep-UV Emission from Highly-Ordered AlGaN/AlN Core-Shell Nanorods

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ABSTRACT: 3D core-shell nanostructures could resolve key problems existing in conventional planar deep-UV LED technology due to their high structural quality, high-quality non-polar growth leading to a reduced quantum-confined Stark effect, and their ability to improve light extraction. Currently, a major hurdle to their implementation in UV-LEDs is the difficulty of growing such nanostructures from $\text{Al}_x\text{Ga}_{1-x}\text{N}$ materials with a bottom-up approach. In this paper, we report the successful fabrication of an AlN/ $\text{Al}_x\text{Ga}_{1-x}\text{N}$ /AlN core-shell structure using an original hybrid top-down/bottom-up approach, thus representing a breakthrough in applying core-shell architecture to deep-UV emission. Various AlN/ $\text{Al}_x\text{Ga}_{1-x}\text{N}$ /AlN core-shell structures were grown on optimized AlN nanorod arrays. These were created using Displacement Talbot Lithography, a two-step dry-wet etching process, and optimised AlN MOVPE regrowth conditions to achieve the facet recovery of straight and smooth AlN non-polar facets, a necessary requirement for subsequent growth. Cathodoluminescence hyperspectral imaging of the emission

characteristics revealed that a 229 nm deep-UV emission was achieved from the highly uniform array of core-shell AlN/Al_xGa_{1-x}N/AlN structures, which represents the shortest wavelength achieved so far with a core-shell architecture. This hybrid top-down/bottom-up approach represents a major advance for the fabrication of deep-UV LEDs based on core-shell nanostructures.

KEYWORDS: nanorod, core-shell, AlN, AlGa_xN, TEM, EDX, Cathodoluminescence

It is well-known that the III-Nitride semiconductors are an important class of materials for devices emitting in the ultraviolet (UV) with applications including: UV curing,¹ medical diagnostics, phototherapy,² optical sensing,³ security, communications,⁴ sterilisation and water and air purification.^{5,6,7} These applications are made possible thanks to the widely tunable bandgap energy of AlGa_xN alloys ranging from 3.4 eV for GaN to 6.2 eV for AlN. The emission wavelength of AlGa_xN-based LEDs can cover the entire UV-A, UV-B, and UV-C spectral range. This represents one of the major advantages compared to the mercury lamps and gas lasers that possess a fixed and limited emission spectrum. Additionally, III-Nitride UV-based LEDs and LDs are compact, robust, environmentally-friendly, offer a long lifetime and have a considerably lower power requirement. However, while visible white-light LEDs based on indium-containing III-Nitrides are today sufficiently efficient and cheap to be established as the leading technology for public and home lighting, UV LEDs still require substantial improvements. Currently, the external quantum efficiency (EQE) of Al_xGa_{1-x}N LEDs barely reach a maximum of 40 % in the UV-A,⁸ 20% in the UV-B,⁹ and up to a few percent in the deep UV-C.^{8,10} This is significantly lower than the 80 % EQE achieved for GaN-based blue LEDs.^{11,12} This gap in terms of efficiency between deep UV and visible LEDs is due to several factors, such as the high density of point and extended

defects, which can act as non-radiative recombination centres in AlN and $\text{Al}_x\text{Ga}_{1-x}\text{N}$ materials,^{13,14} the low carrier injection and difficulties in achieving efficient p-doping in AlN-rich $\text{Al}_x\text{Ga}_{1-x}\text{N}$ alloys,^{15,16} the poor light extraction,^{17,18,19} and the internal electric field in quantum wells,^{20,21} which are exacerbated compared to GaN due to the much larger spontaneous polarisation properties of AlN-based materials.²²

In a large part, these challenges result directly from the approach employed to fabricate these LEDs, which is nowadays based on a planar technology. The use of 3D nanostructures is an approach to address and mitigate some of these challenges. In particular, the use of nanorod arrays can allow the reduction of extended defects compared to those in the initial planar template by means of dislocation bending²³ or filtering,^{24,25} thus reducing the threading dislocations density in the active region. Furthermore, a core-shell nanostructure configuration, with the core along the c -direction, involves radial growth of the quantum wells (QWs) on non-polar facets where strong spontaneous and piezoelectric polarisation are absent.²⁶ The mitigation of the quantum-confined Stark effect (QCSE) will improve the overlap between electron and hole wave functions and thus the emission efficiency. Additionally, high densities of basal-plane stacking faults (BSFs) have been observed in planar non-polar AlGaIn layers,²⁷ since BSFs can effectively compensate lattice translations between islands that occur during the early stage of heteroepitaxial growth.²⁸ In contrast, nanostructures aligned along the c -direction are formed from growth in the polar direction, for which BSFs are not observed away from the substrate interface. Therefore, the non-polar sidewalls of core-shell nanostructures offer the prospect of a higher structural quality for the QWs, and hence an increase in the internal quantum efficiency (IQE). Moreover, the QW growth, occurring radially to the nanorod core, can dramatically increase the overall emitting area over that of a planar equivalent.²⁹ Such a benefit is important because poor efficiency in nitride LEDs at

high current injection occurs at high carrier densities. Spreading a high current over the larger emissive surface area of core-shell structures brings the device back into its high-efficiency, low-carrier density regime.³⁰ Finally, the use of nanostructures also improves the light extraction efficiency (LEE) of the devices, for example by randomising the angular distribution of the photons in the LED.^{31,32}

However, a key obstacle to the implementation of core-shell structures in UV devices is the difficulty of growing such regular structures in $\text{Al}_x\text{Ga}_{1-x}\text{N}$ -based materials. Whereas the selective area growth (SAG) of GaN nanorod arrays and subsequent InGaN/GaN based core-shell LEDs has been demonstrated by metal organic vapour phase epitaxy (MOVPE),^{33,34,35,36} the SAG of AlN and Al-rich AlGaIn nanorods has not been achieved yet due to the very high sticking coefficient and the low diffusion length of Al atoms.^{37,38} A recent report of AlGaIn nanorods by molecular beam epitaxy (MBE) on a Si substrate,³⁹ shows how AlGaIn nanostructures could be used to circumvent the challenges of planar technology. However, they require a GaN pedestal grown on Si to initiate the nucleation.^{39,40,41} Ultimately, sapphire substrates and GaN-free structures are required for devices operating in the UV-C regime to avoid absorption of the UV-C light. Additionally, MBE-based nanorods have, up to now, only been demonstrated to be suitable for the growth of axial structures and not core-shell structures, which provide larger active areas. The recent report of Kim et al. represents the only success to date to create an AlGaIn/AlN core-shell structure.⁴² They report the fabrication of AlN nanorods on a sapphire substrate by polarity-selective epitaxy and etching (PSEE) and achieved a 260 nm emission from core-shell MQWs. Their results further highlight the potential of the core-shell geometry with an improved PL intensity and IQE of the MQW over one grown on planar AlN. However, their approach is based on the use of Al-polar AlN inversion domains to create the core, which leads to irregular and unorganised structures; unsuitable for real

devices. An alternative route, already demonstrated for InGaN/GaN core-shell nanorod arrays,^{43,44,45} uses top-down etching followed by MOVPE regrowth. It is a promising route for the fabrication of uniform and highly organised AlN nanorod arrays that will subsequently allow the creation of AlGaN/AlN core-shell devices.^{46,47} We have recently demonstrated this first critical step prior to creating device structures with active quantum wells.³⁸

In this paper, we present uniform and organised core-shell AlN/AlGaN/AlN nanorod arrays with a vertical nonpolar AlGaN/AlN single QW, using this hybrid top-down/bottom-up approach. The emission characteristics of the AlGaN/AlN QW are studied in detail by cathodoluminescence hyperspectral imaging. Uniform emission from the non-polar facets is tuned to cover the UV-C spectral range from 265 nm to 229 nm, which represents the shortest wavelength achieved so far with a core-shell architecture. The results show the potential of the hybrid top-down/bottom-up approach to achieve nonpolar core-shell AlGaN/AlN nanorod arrays on a wafer scale to achieve deep-UV light emitting devices.

RESULTS AND DISCUSSION

The AlGaN/AlN nonpolar core-shell nanorod array was fabricated using a hybrid approach composed of top-down etching and subsequent bottom-up MOVPE regrowth. To create the AlN nanorod cores, a $\sim 4\text{-}5\text{ }\mu\text{m}$ AlN template grown by MOVPE on (0001) sapphire substrates was employed. We used Displacement Talbot Lithography (DTL) to fabricate a metal dot mask via a lift-off process, in a similar way to our previous work.^{38,47} A detailed description of the DTL patterning and lift-off process is presented in the Methods section. This fast and robust fabrication

process, performed on a full 2-inch wafer, allows the creation of a hexagonal array of metal dots with a diameter of ~ 250 nm and a pitch of $1.5\ \mu\text{m}$, as shown in Figure S2.

Inductively coupled plasma (ICP) etching with a chlorine chemistry was then used to transfer the metal mask into the 2-inch AlN template, following the optimum etching conditions presented in our previous work and detailed in the Methods section.^{38,47} A potassium hydroxide (KOH) based wet etching was then applied by soaking the wafer in AZ 400K at $20\ ^\circ\text{C}$. Figure 1.a shows an SEM image of the resulting smooth and straight sidewalls created after both the dry and wet etching processes. This is further evidenced by TEM observation on scratched AlN etched nanorods in Figure 2.a and S4. AlN nanorods with a diameter of ~ 130 nm and a height of $1.8\ \mu\text{m}$ were achieved. Note, the wet etching step reduces the nanorod diameter from that obtained after dry etching (Figure S3). AZ 400K solution was chosen as an etchant due to its low concentration of KOH, allowing a better control of the structure shape and etch rate. Note that similar wet etch chemistry has been used to control the dimensions of dry etched GaN nanorods,⁴⁸ although AlN and GaN have different chemical reactivity.

Subsequently, the AlN nanorod scaffold was placed in a MOVPE reactor for the regrowth of AlN facets, at $1100\ ^\circ\text{C}$, 20 mbar, a V/III ratio of 30554 and with H_2 as the carrier gas.^{38,47} More details are given in the Methods section. Figure 1.b demonstrates that AlN regrowth has been achieved along the whole height of the nanorod. Well-defined *m*-plane facets are formed along the nanorod sidewall together with a truncated pyramid on the top composed of six (10-11) semi-polar planes and a top *c*-plane. In contrast to previous reports of AlN regrowth on etched nanorods,^{46,47} straight and smooth non-polar sidewall facets are achieved. Further evidence from TEM observations on scratched AlN faceted nanorods can be found in Figure 2.b and S5. An *m*-plane growth rate of $\sim 1\ \text{nm/min}$ was extracted from SEM and TEM observations before and after

regrowth.

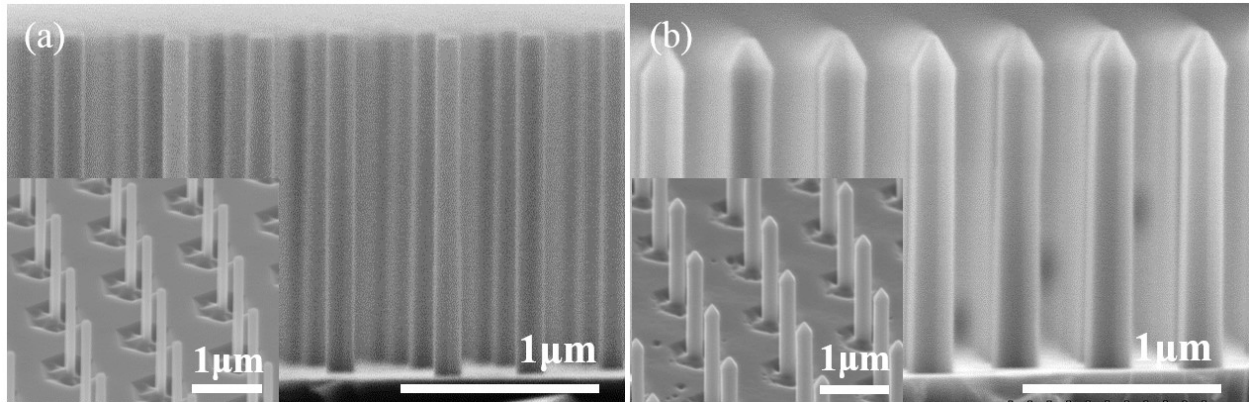


Figure 1. Cross-section SEM images of (a) AlN nanorod arrays after ICP dry etching and AZ400K wet etching and (b) AlN faceted nanorod arrays after MOVPE regrowth. The inset in (a) and (b) show a tilted-view of the AlN etched and faceted nanorods, respectively.

This significant advance results from the choice of AlN regrowth conditions and the initial morphology and configuration of the AlN nanorod array after the two-step etching process. Firstly, the formation of smooth and straight facets is enhanced at low pressure and high V/III ratio.⁴⁷ Low pressure enhances Al-adatom diffusion, allowing a complete coverage of the AlN nanorod. High V/III ratio reduces the growth rates of both the *c*- and *m*-planes relative to the *a*-plane growth rate, favouring a complete coverage of the AlN nanorod, but also mitigating the growth on the top pyramidal part, which is not favourable for a smooth and straight facet regrowth.^{46,47} Secondly, the use of straight and smooth AlN etched nanorods prevents the formation of regrowth steps that can be induced by etch damage, especially in these growth conditions where the *m*-plane growth rate is lowered.⁴⁷ The small nanorod diameters used in this work allows the formation of *m*-plane facets along the whole height of the nanorod, which is not the case for larger diameters.⁴⁷ Finally, the use of a relatively large 1.5 μm pitch allows good gas-phase diffusion of the growth reagents to

the base of the nanorods, which ensures that there is no variation in the m -plane growth rate along the height of the nanorod.⁴⁶

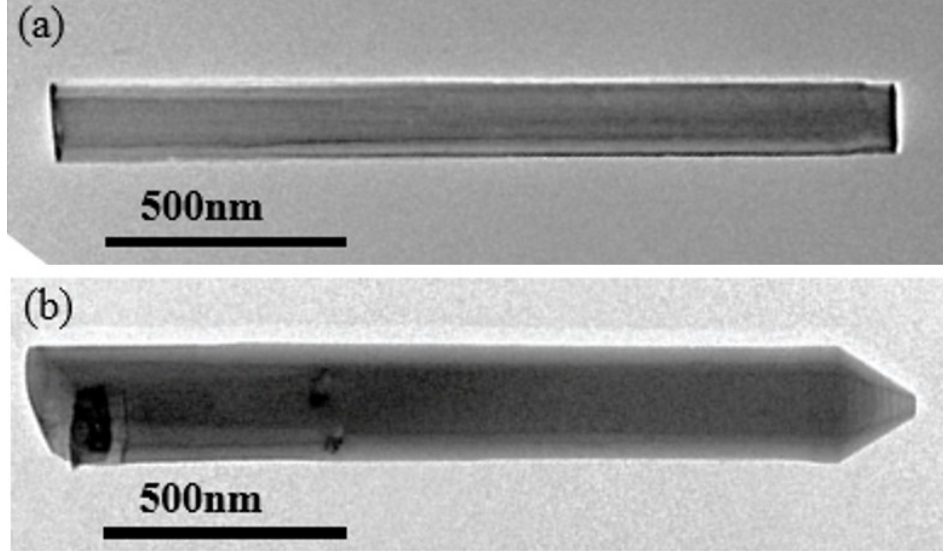


Figure 2. TEM images of scratched (a) AlN nanorod arrays after ICP dry etching and AZ400K wet etching and (b) AlN faceted nanorod arrays after MOVPE regrowth.

Following the AlN faceting regrowth step, an AlGa_{0.1}N shell was created by adding 2 sccm of TMGa for 2 minutes and setting the NH₃ flow rate to 500 sccm ($V/III = 1638$), with otherwise the same growth conditions, resulting in an approximate thickness of ~ 2 -3 nm based on the m -plane AlN growth rate previously extracted. An AlN cap layer was then grown for 15 minutes with the same AlN conditions to create an AlGa_{0.1}N/AlN SQW (AlGa_{0.1}N SQW 1). The resulting core-shell structures are displayed in Figure 3a and 3b. Both SEM and TEM images show straight and smooth sidewalls and a fully-formed nanopyramid on the top. No visible extended defects, such as threading dislocations (TDs) or basal stacking faults (BSFs), are observed in the nanorod shown in Figure 3.b. The creation of the 130 nm diameter AlN nanorod array has acted to filter the TDs, as shown in Figure 2.a and S4, so that zero or at the most one TD is observed in the resulting

nanorods after AlN regrowth.^{24,25} Any TDs present often bend towards the lateral free surfaces and can be seen as surface pits in Figure 2.b and Figure S5. Figures 3c, 3d and 3e display the aluminium, nitrogen and gallium concentration, respectively, of the grown core-shell structures as measured by EDX spectroscopy. Gallium incorporation is observed across the whole height of the nanorod as well as on the semi-polar plane.

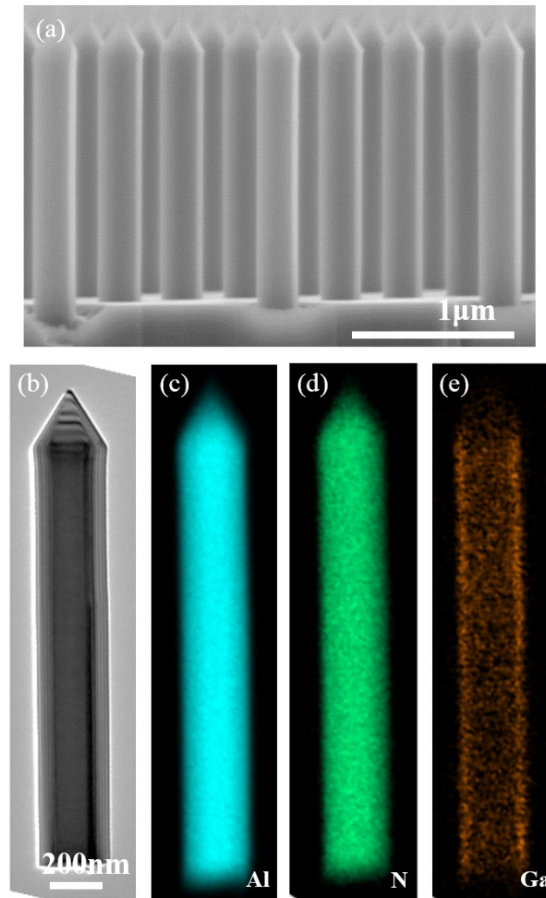


Figure 3. (a) Cross-section SEM and (b) TEM pictures of AlN/AlGaIn SQW core-shell structures (AlGaIn SQW 1) grown by MOVPE. (c) Al, (d) N and (e) Ga concentration maps obtained by EDX spectroscopy.

The optical emission of the AlN/AlGa_N/AlN core-shell structures was assessed by room-temperature high-resolution cathodoluminescence (CL) hyperspectral imaging in a low vacuum SEM (ESEM) and is presented in Figure 4. The intensity map in Figure 4.b shows the extracted emission intensity of the SQW between 4.9-5.5 eV for a single AlN/AlGa_N/AlN core-shell nanorod (Figure 4.a). Higher emission intensity arises from the top of the nanorod, at the corners between the semi-polar and non-polar planes, and where regrowth has taken place on the etched planar AlN surface between the nanorods. Indeed, after the two etching steps, hexagonal trenches are created at the base of the nanorods (inset in Figure 1.a). While these do not impact the AlN faceting regrowth step, preferential incorporation seems to occur during the SQW growth. This “parasitic growth” could be suppressed by passivating the AlN bottom *c*-plane with a hydrogen silsesquioxane layer.^{Error! Bookmark not defined.}

Focusing on the nanorod emission characteristics, Figure 4.c shows a line spectrum extending from top to bottom of the nanorod (excluding the parasitic emission), and displayed on a log scale. Both the AlN near band edge (NBE) emission and AlGa_N SQW emission can be observed along the whole height of the nanorod. The broad AlN NBE emission at 5.8-6.0 eV (207-214 nm) displays a higher intensity on the top semi-polar part of the nanorod. The AlN NBE emission range agrees with that reported in the literature at room temperature.^{49,50,51} A lower energy broad emission band is generally observed between 2.6 and 4.3 eV for all samples (Figure S6 and S7). The high intensity of the defect luminescence is partly due to the low vacuum mode in which the CL images were acquired and is not indicative of the quality of the sample (see Supporting Information for further discussion). The defect band, initially observed in the AlN template,⁴⁷ could be related to O-complexes and/or Si-complexes and other native defects, such as vacancies.^{38,47,52,53} The AlGa_N SQW shows two emission energies: one centred around 5.13 eV (242 nm) on

both the *m*-plane facets and semi-polar facets; and a second one centred around 5.22 eV (238 nm) having a higher intensity and localised at the corners between the semi-polar and non-polar nanorod facets. The small energy shift occurring at the corners between the semi-polar and non-polar planes could be due to a change of the surface morphology at the intersection, preferential incorporation of species as a result of strain relaxation at the corners, or from a local change of the QW thickness, similar to that observed for InGaN/GaN core-shell structures.^{54,55} Note that the higher intensity could also result from a local change to the QW thickness or enhanced light extraction.

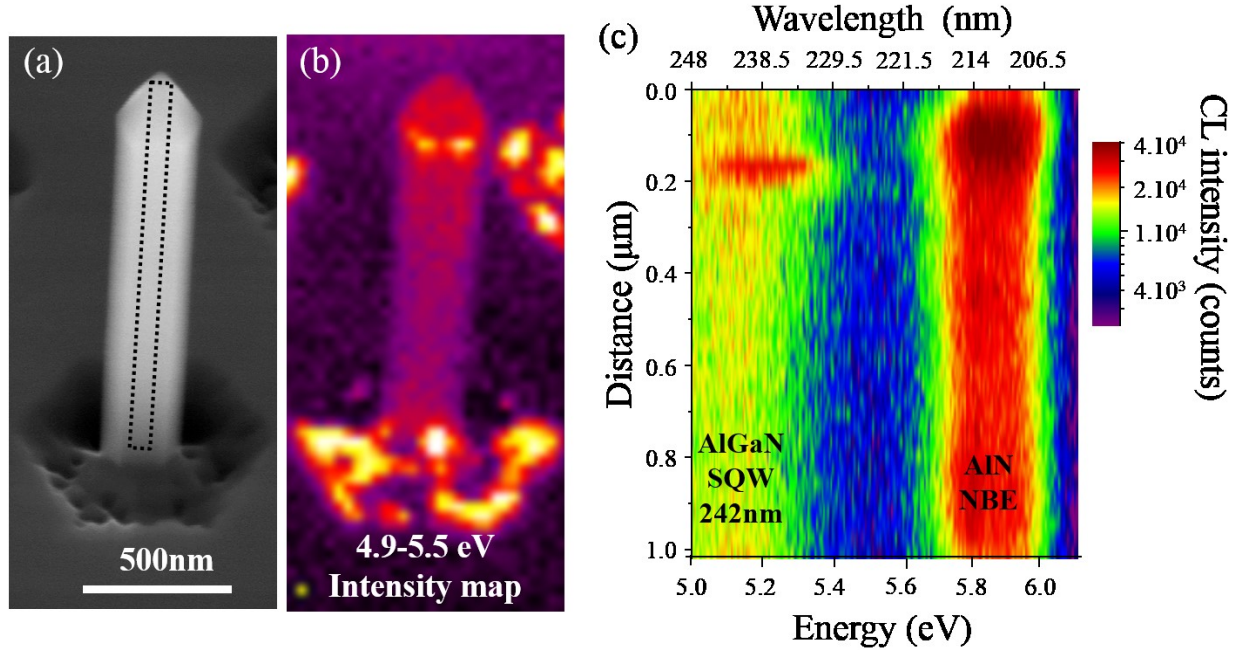


Figure 4. (a) Tilted SEM images of AlGaIn SQW 1. (b) Related intensity map extracted over the SQW emission range of 4.9-5.5 eV. (c) Log-scale RT spectral line-spectrum extracted along the length of the nanorod displayed in (a) from the region highlighted by the dashed rectangle.

Based on the results presented in Figure 4, additional AlGa_N/AlN SQW growths have been performed in order to improve the emission characteristics, such as intensity and uniformity, but also to tune the emission wavelength. Figure 5.a, b and c display the characteristics obtained at lower growth temperature and higher pressure (AlGa_N SQW 2) while Figure 5.d, e and f shows the characteristics for an additional increase in TMGa flow rate (AlGa_N SQW 3). The SQW thickness can be approximately estimated to be ~ 2-3 nm and ~ 10-15 nm respectively for AlGa_N SQW 2 and 3. More details on the SQW growth conditions are given in the Methods section. Compared to AlGa_N SQW 1 (Figure 4.b), the intensity map extracted for AlGa_N SQW 2 between 5-5.7 eV (Figure 5.b) suggests an improved intensity of the emission from the nanorod sidewalls to that from the planar *c*-plane. The line spectrum extracted from the top to the bottom of the nanorod in Figure 5.c confirms the improvement of the AlGa_N SQW emission centered around 229 nm (5.41 eV): it has a similar intensity to the AlN NBE, and a good emission uniformity across the whole height of the nanorod. Utilising a lower growth temperature and higher pressure likely decreased the Al-adatom diffusion, and hence mitigated any preferential incorporation on strain-relaxed surfaces. The lower emission wavelength of the SQW can be explained by Ga desorption during the ramp in temperature between the SQW growth and the AlN cap layer growth.

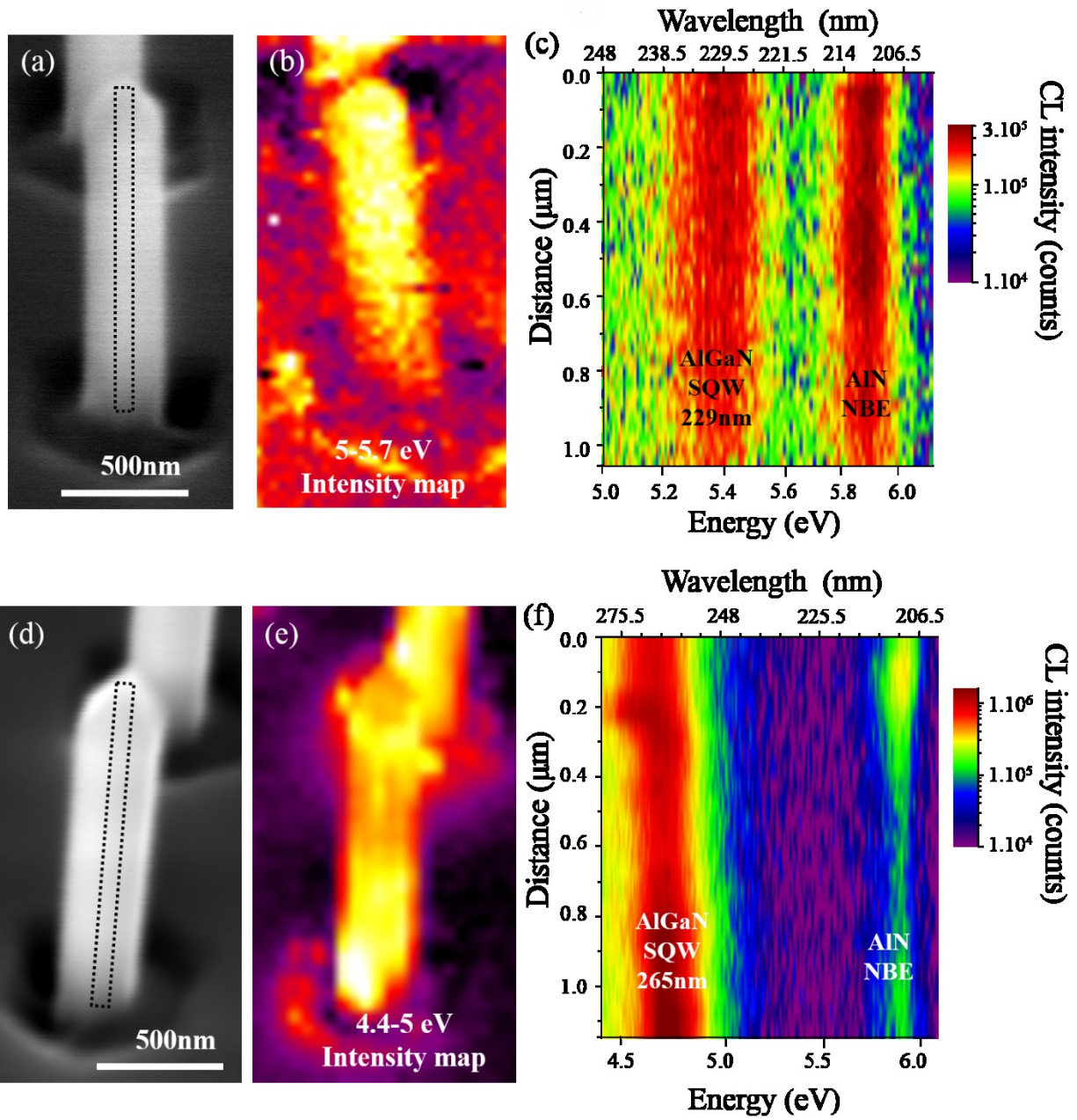


Figure 5. (a) Tilted SEM images of AlGaIn SQW 2. (b) Corresponding intensity map extracted over the SQW emission range of 5-5.7 eV. (c) Log-scale RT spectral line-spectrum extracted along the length of the nanorod displayed in (a) from the region highlighted by the black dashed rectangle. (d) Tilted SEM images of AlGaIn SQW 3. (e) Corresponding intensity map extracted

over the SQW emission range of 4.4-5 eV. (f) Log-scale RT spectral line-spectrum extracted along the length of the nanorod display in (d) and from a region highlighted by the dashed rectangle.

The increase in TMGa flow rate by a factor of five (AlGaIn SQW 3), while keeping all other growth conditions constant, results in a clear red-shift of the emission energy range of the SQW, from 5-5.7 eV to 4.4-5 eV (Figure 5.e). The intensity is no longer uniform (Figure 5.e) with some small differences observed from different parts of the nanorod, with higher intensities at the intersections between the semi-polar and non-polar planes, at intersections between two non-polar planes (vertical line along the height of the nanorod) and at the bottom of the nanorod. The emission along the height of the nanorod is also no longer uniform with a slight red-shift in emission energy from the bottom to the top of the nanorod. The five-fold increase in TMGa, while keeping the same growth time, is expected to lower the emission energy and increase the growth rate; thus creating a thicker AlGaIn layer. As already reported for InGaIn/GaN core-shell nanorods,^{56,57} a small thickness gradient along the height of the nanorod can lead to a gradual shift of the emission. Additionally, the observation of a higher intensity at the intersections of the planes can also arise from the growth of a thicker layer since it leads to preferential incorporation of species on strain-relaxed regions.^{54,55} This preferential incorporation can lead to the formation of a quantum-wire-like structure that will enhance the recombination rate and hence the intensity.

Regardless of the small non-uniformity of the emission observed for AlGaIn SQW 3, the overall SQW intensity is drastically improved for growth conditions of AlGaIn SQW 3. Indeed, while the AlN NBE emission remains more or less at the same intensity ($\sim 10^5$ counts) between AlGaIn SQW 2 and 3 (respectively, Figure 5c and 5f), the AlGaIn SQW intensity is increased by an order of magnitude in AlGaIn SQW 3. Similarly, significant improvement in the nanorod intensity relative to the planar *c*-plane is also observed. This can be clearly observed in Figure 6 which compares

for the three AlGaIn SQWs the spectra acquired on the *m*-plane nanorod sidewalls (full line) with the one acquired on the planar un-passivated *c*-plane (dashed line). While the intensity is increased by a factor 1.5 for the SQWs emitting at 242 nm and 229 nm (respectively AlGaIn SQW 1 and 2), the enhancement factor reaches 5 for the SQW emitting at 265 nm (AlGaIn SQW 3). For all AlGaIn SQWs, the enhancement could be attributed to the higher structural quality of the AlN nanorod template compared to the *c*-plane in between, and to a better overlap between the electron and hole wave functions within the non-polar SQW. In addition, the polarisation dependence on substrate orientation should be considered. For *c*-plane growth, polarisation switching from light with a dominant polarisation of $E \perp c$ to light with a dominant polarisation of $E \parallel c$ occurs when the Al concentration increases. The switch occurs between $x \approx 0.25$ (~ 300 nm emission)^{58,59} and $x \approx 0.8$ (~ 230 nm)⁶⁰ depending on the strain in the QW, its thickness and the internal electric fields.^{60,61} For *m*-plane growth, the change in the SQW symmetry results in a modification of the valence subbands which leads to the lowest energy transition being strongly polarised along $E \parallel c$ for a wide range of Al compositions.^{62,63} Hence, the emission is likely to be polarised along $E \parallel c$ for both the bottom *c*-plane and lateral nanorod *m*-planes, with the latter orientation resulting in less reabsorption due to reduced guiding in the plane of the QW. Further improvement for AlGaIn SQW 3 could be due to higher carrier confinement and/or reduced carrier leakage due to the thicker QW. These results demonstrate that optimising the growth conditions enables tuning of the emission wavelength and significantly improves the intensity while maintaining a high or relatively high emission uniformity from the whole height of the nanorods.

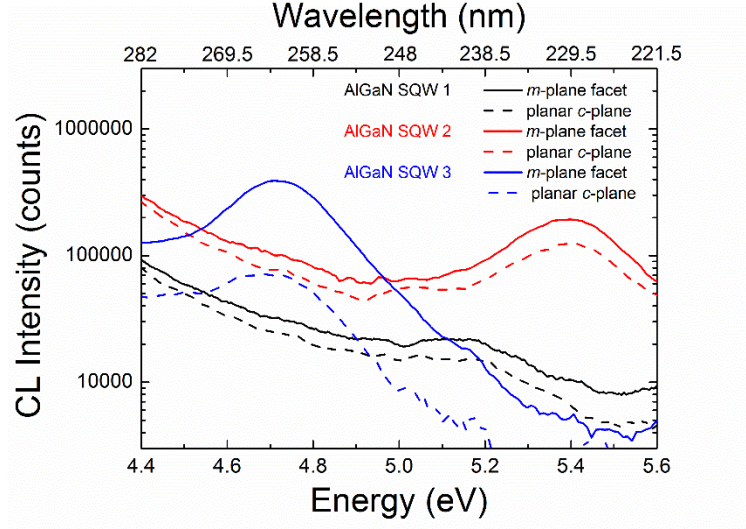


Figure 6. CL spectra acquired for the three AlGaIn SQWs showing both emission from the *m*-plane nanorod sidewalls (full line) and also from the *c*-plane material in between the nanorods (dashed line).

A striking result that is observed in all the AlGaIn SQWs relates to the peak emission wavelength coming from the different planes. As observed in the line spectra extracted along the height of the nanorods in Figure 5.c and 5.f and on the CL spectra acquired on both *c*-planes and *m*-planes in Figure 6, no difference in the peak wavelength was observed between different planes. The resulting high uniformity of the emission of the overall core-shell structures, irrespectively of the plane orientation, represents a major advantage compared to visible InGaIn/GaN core-shell structures where the indium incorporation is highly sensitive to the plane orientation.^{64,65} For visible InGaIn/GaN core-shell structures this generally induces a change of the EL peak emission position as a function of the injection current.³⁶

It should be noted that although several electrically-injected InGaIn/GaN core-shell LEDs emitting in the visible have been demonstrated, there are still further obstacles before devices based on AlGaIn/AlN core-shell structures are realised. One major issue is the poor n-type conductivity

of the AlN layer forming the core. This could be circumvented by the use of mid to high Al content n-type AlGaIn layer for which relatively good carrier density has been reported.⁶⁶ Since the hybrid top-down/bottom-up approach can be transferred to any III-nitride template, both binary and ternary alloys, its application to state of the art n-type AlGaIn layers represents the most promising solution for electrically-injected core-shell structures emitting in the deep UV.

CONCLUSION

The paper demonstrates an original and reproducible approach for creating highly uniform and organised arrays of core-shell AlN/AlGaIn/AlN nanorods that avoids the difficulties of creating bottom up nanostructures in the AlGaIn materials system. In the combined top-down-etching/bottom-up-growth approach, we employ the recently-developed nanolithographic technique of Displacement Talbot Lithography to create a metal dot etch mask across a full 2-inch wafer via lift-off. The successful fabrication of uniform AlN nanorods with smooth vertical facets is then achieved by the sequential use of dry etching, wet etching and MOVPE regrowth using optimised conditions. This led to large areas of uniform nanorods with an aspect ratio as high as 12. A future reduction of the nanorod pitch from 1.5 to 1 μm with the same nanorod aspect ratio, which we believe is technically feasible with this technique, would lead to a two-times enhancement of the active area compared with planar QWs; thus potentially allowing more efficient LED operation by mitigating droop.

An AlGaIn single quantum well has been successfully regrown on the AlN nanorods along their entire height, as determined by detailed electron-beam spectroscopic measurements, with a significant improvement of the emission coming from the non-polar facets compared to the *c*-plane planar growth. Deep UV emission at 229 nm is reported on AlN/AlGaIn/AlN core-shell structures for the first time. No facet-dependent alloy incorporation is observed, in contrast to visible-

emitting core-shell structures, which will simplify the implementation of these 3D structures into electrically-driven devices by allowing a more stable emission wavelength with drive current.

The approach for creating nanostructures that combines top-down-etching with bottom-up growth shows great promise for the fabrication of deep-UV LEDs based on core-shell nanostructures due to the flexibility of creating cores with a wide range of $\text{Al}_x\text{Ga}_{1-x}\text{N}$ alloy compositions and the controllability of creating regular arrays with a high degree of diameter and height uniformity. Such LEDs will have application, for example, in water and air sterilisation.

METHODS

Displacement Talbot Lithography Patterning. Commercial AlN templates (Nanowin) were spin-coated at 3000 rpm with a bottom antireflective layer (BARC) (Wide 30C, Brewer Science) to obtain a layer thickness of 290 nm, followed by a 240 nm layer of positive photoresist (Ultra-i 123, Dow Electronic Materials). We used Displacement Talbot lithography (DTL) (PhableR 100, Eulitha) to expose the photoresist with a ~ 375 nm laser source through a conventional photomask comprising 800-diameter holes in a hexagonal arrangement with a $1.5\ \mu\text{m}$ pitch. The initial proximity gap was set to $150\ \mu\text{m}$ and the displacement during exposure was $71.2\ \mu\text{m}$, corresponding to eight Talbot lengths. After exposure, a post-exposure bake for 1 min 30 sec at $120\ ^\circ\text{C}$ was applied before development in MF-CD-26. Figure S1 shows the 260 nm diameter openings achieved after the DTL patterning process.

AlN Nanorod Array Fabrication. An electron-beam evaporator was used to deposit 10 nm of Au and 200 nm of Ni on the AlN wafer to create metal masks in the circular opening at the AlN surface. The positive resist and wet-developable BARC was used to create a lift-off profile with MF-CD-26 developer and the AlN wafer cleaned in a 2 min reactive-ion etching (RIE) oxygen

plasma to remove any BARC residue prior to evaporation. The resulting ~250 nm Au/Ni dots after lift-off are displayed in Figure S2. AlN nanorods were then created using an inductively coupled plasma (ICP) dry etcher (Oxford Instruments System 100 Cobra) using conditions reported previously.³⁸ Any passivation layer on the slightly undercut nanorod sidewalls (Figure S3.a) was then removed in buffered oxide etchant prior to further wet-etching in AZ400K potassium-hydroxide-(KOH)-based developer to further smooth and shrink the diameters of the nanorods (Figure 1.a and S3.b). Finally, the Ni mask was removed in HCl:HNO₃, 3:1.

Faceting regrowth of AlN nanorods by MOVPE. The bottom-up regrowth of AlN and AlGaIn was carried out in a 1 x 2” horizontal Aixtron MOVPE reactor. The employed growth parameters are summarised in Table 1. The growth conditions to satisfy a smooth and straight AlN faceting regrowth follow our previous report,³⁸ and summarized here: a growth temperature of 1100 °C, a pressure of 20 mbar, 10 sccm in TMAI flow rate, 4000 sccm in NH₃ flow rate (V/III ratio of 30554) and H₂ as the carrier gas. AlGaIn single quantum well (SQW) growth was performed for three different conditions: for AlGaIn SQW 1, depicted in Figure 3 and 4, a growth temperature of 1100 °C, a pressure of 20 mbar, 10 sccm in TMAI flow rate, 2 sccm in TMGa flow rate, 500 sccm in NH₃ flow rate (V/III ratio of 1638) and H₂ as the carrier gas; for AlGaIn SQW 2, depicted in Figure 5a, b and c, a growth temperature of 1050 °C, a pressure of 100 mbar, 10 sccm in TMAI flow rate, 2 sccm in TMGa flow rate, 500 sccm in NH₃ flow rate (V/III ratio of 1638) and H₂ as the carrier gas; for AlGaIn SQW 3, depicted in Figure 5d, e and f, a growth temperature of 1050 °C, a pressure of 100 mbar, 10 sccm in TMAI flow rate, 10 sccm in TMGa flow rate, 500 sccm in NH₃ flow rate (V/III ratio of 499) and H₂ as the carrier gas. The AlN cap layer was grown with the same conditions as the AlN faceting regrowth step.

<i>Growth step</i>	<i>Temperature</i>	<i>Pressure</i>	<i>NH₃</i>	<i>TMAI</i>	<i>TMGa</i>	<i>V/III ratio</i>
	(°C)	(mbar)	(sccm)	(sccm)	(sccm)	
<i>AlN faceting</i>	1100	20	4000	10	x	30554
<i>AlGaN SQW 1</i>	1100	20	500	10	2	1638
<i>AlGaN SQW 2</i>	1050	100	500	10	2	1638
<i>AlGaN SQW 3</i>	1050	100	500	10	10	499
<i>AlN cap</i>	1100	20	4000	10	x	30554

Table 1. Growth parameters employed for the different samples

Structural Characterisation. Scanning electron microscopy (SEM) was used to monitor the fabrication process and determine the morphology and dimensions of the structures, using a Hitachi S-4300 SEM. The morphology and chemical information of AlGaN/AlN core-shell structures were investigated using a JEOL 2100 200 kV TEM. The core-shell nanorods used for TEM observations were mechanically removed from the wafer and deposited on a carbon grid, hence no mechanical-polishing or focused ion beam thinning has been performed.

Optical Characterisation. Cathodoluminescence hyperspectral imaging measurements were carried out on the AlGaN/AlN core-shell nanorods at room temperature in a modified FEI Quanta 250 field emission scanning electron microscope. The electron beam energy was set to 15.0 keV and the beam current to ~7 nA. A reflecting objective (NA 0.28) with its axis perpendicular to the electron beam collected the light and focused it on the 50 µm entrance slit of the spectrograph

using an off-axis paraboloidal mirror. The spectrograph had a focal length of 125 mm, and the diffracted light from the 600 lines/mm grating was focused onto a cooled electron-multiplying charge-coupled device (EMCCD) detector.

ASSOCIATED CONTENT

The Supporting Information is available free of charge on the ACS Publications website at DOI: Detail on DTL patterning and lift-off to create a hard etch mask and additional SEM images; SEM images comparing nanorod after dry and wet etch; additional TEM images of etched and faceted nanorods; further results and details of the cathodoluminescence characterisation.

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Author Contributions

P.-M.C. conceived the experimental work, supervised by P.A.S. P.-M.C. carried out the nanorod fabrication, MOVPE growth experiment, and SEM characterisation. G. K and R.W.M. performed the CL characterisation. All the authors contributed in analysing and writing the results.

Notes

The authors declare no competing financial interest.

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